Energy, Mines and Resources Canada | Énergie, Mines et Ressources Canada

CANMET

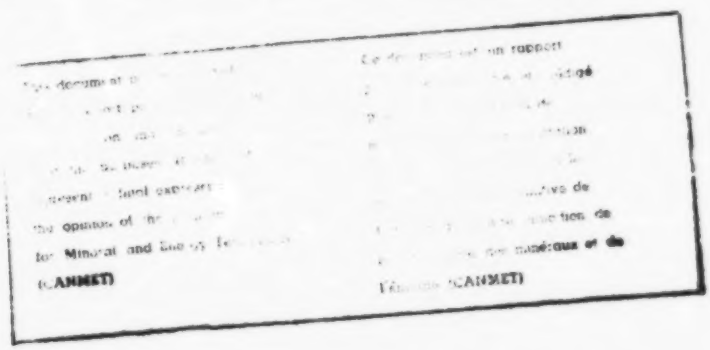
Canada Centre for Mineral and Energy Technology | Centre canadien de la technologie des minéraux et de l'énergie

ENERGY DEPENDENCE OF ULTRA-THIN $CaSO_4$ ·Tm CHIPS TO RADON AND THORON PROGENY

H. LI and J. BIGU

ELLIOT LAKE LABORATORY

NOVEMBER 1986



17pp

MICROMEDIA

MINERAL AND ENERGY TECHNOLOGY
MINING RESEARCH LABORATORIES
DIVISION REPORT M&ET/MRL 86-158(TR)

ENERGY DEPENDENCE OF ULTRA-THIN $CaSO_4$·Tm CHIPS TO RADON AND THORON PROGENY

by

H. Li* and J. Bigu**

ABSTRACT

The response of ultra-thin (2 mg/cm^2) $CaSO_4$·Tm thermoluminescent (TLD) chips to α-particles from radon and thoron progeny has been investigated. The energy of the α-particles was changed by adding polycarbonate absorbers of different thickness, i.e., up to 46 μm, to radon progeny or thoron progeny deposited on 0.8 μm Millipore filters. It is shown that the sensitivity of the TLD material is energy-dependent. However, for filters with no absorbers, the energy dependence was minimal and the sensitivity of ultra-thin $CaSO_4$·Tm can be taken as the same for radon progeny and thoron progeny. This experimental finding considerably simplifies the calibration of α-particle personal dosimeters using the above TLD material for field applications in radon/thoron atmospheres such as those encountered in some underground uranium mines.

Keywords: Thermoluminescent detector, personal dosimeter, TLD chip, radon progeny, thoron progeny.

* Radiation Protection Engineer, Uranium Mining Research Institute, Hengyang (China), and 1986 Fellow of International Atomic Energy Agency (IAEA), Vienna (Austria).

** Research Scientist and Radiation/Respirable Dust/Ventilation Project Leader, Elliot Lake Laboratory, CANMET, Energy, Mines and Resources Canada, Elliot Lake, Ontario.

DÉPENDANCE ÉNERGÉTIQUE DES PASTILLES ULTRA-MINCES $CaSO_4$:Tm ENVERS LES DESCENDANTS DU RADON ET DU THORON

par

H. li* et J. Bigu**

RÉSUMÉ

On a étudié la réaction des pastilles ultra-minces (2 mg/cm^2) de $CaSO_4$:Tm thermoluminescentes (TLD) envers les particules alpha provenant des descendants du radon et du thoron. L'énergie des particules alpha a été changée en ajoutant des absorbeurs de polycarbonate d'épaisseurs différentes, c.-à-d. jusqu'à 46 µm, aux descendants du radon ou du thoron déposés sur des filtres Millipore de 0,8 µm. Au cours de l'étude, la sensibilité des matériaux LTD, a démontré une dépendance énergétique. Toutefois, chez les filtres absorbeurs, la dépendance énergétique était minime alors que la sensibilité des pastilles ultra-minces $CaSO_4$:Tm était la même pour les descendants du radon et du thoron. Cette découverte simplifie considérablement l'étallonnage des dosimètres alpha personnels utilisant les matériaux TLD susmentionnés pour des applications sur le terrain dans des atmosphères radon/thoron comme celles que l'on rencontre dans les mines d'uranium souterraines.

Mots-clés: Détecteur thermonumilescent, dosimètre personnel, pastilles TLD, descendants du radon, descendants du thoron

*Ingénieur, protection contre les rayonnements, Institut de recherche minière sur l'uranium, Hengyang (Chine) et membre en 1986 de L'Agence international de l'énergie atomique (AIEA), Vienne, Autriche.

**Chercheur scientifique et chef de projet Rayonnements/Poussières respirables/ Ventilation, Laboratoires d'Elliot Lake, CANMET, Énergie, Mines et Ressources Canada, Elliot Lake, Ontario.

CONTENTS

	Page
ABSTRACT	i
RESUME	iii
INTRODUCTION	1
THEORETICAL BACKGROUND	2
EXPERIMENTAL PROCEDURE	4
RESULTS AND DISCUSSION	9
CONCLUSIONS	12
ACKNOWLEDGEMENT	12
REFERENCES	12

TABLES

No.		
1.	Some theoretical data of radon and thoron progeny	3
2.	TLD response of ultra-thin $CaSO_4$·Tm chip to α-particles of different energy	10

FIGURES

1.	Cross-section of the dosimeter detector head	5
2.	Filters placed in the standard to collect radon or thoron progeny products	5
3.	Four different energy groups of α-particles in determining energy dependence of TL response of ultra-thin CaSO·Tm chips	7
4.	Different incident energy of α-particles emitted by ThC, RaC' and ThC deposited on the filters covered with P.C. absorber with different thickness	8

INTRODUCTION

Methods and techniques using thermoluminescent detectors (TLD) for determining individual radon daughters exposure have been developed for many years and several TLD radon daughters personal dosimeters have been tested in underground uranium mines for technical evaluation purposes (1-7). Because of high sensitivity to γ-radiation, most TLD chips used in α-particle dosimeters, e.g., LiF and CaF_2.Dy, have a lower than desired α-sensitivity to γ-sensitivity ratio, which results in low accuracy and somewhat unsuitable minimum detection limits for some personal dosimetry applications.

A way of increasing the α-particle to γ-radiation sensitivity ratio of a TLD chip is by reducing its thickness to a minimum compatible with the radiation properties of matter, and hence dictated by the interaction characteristics of α- and γ-radiation with the TLD material. (It should be notedthat, although to a lesser extent, the above also applies to β-radiation.)

An 'optimized' thickness compatible with the mechanical properties of the TLD material can be estimated based on the high energy losses of α-particles, and low energy losses of γ-radiation, in traversing matter. However, the mechanical properties of TLD materials are different and it is not always possible and/or practical to reduce the thickness of a given material to the desired amount. For this, and other reasons, TLD chips other than the conventional LiF and CaF_2.Dy chips used for α-particle dosimetry purposes have been investigated for maximum α-particle sensitivity in the presence of gamma fields.

This report presents data on the properties of an ultra-thin TLD chip suitable for high-sensitivity α-particle personal dosimetry purposes. The TLD chip used in this experiment is an ultra-thin $CaSO_4$:Tm chip of 10 mm diameter

and 2 mg/cm^2 sensitive layer thickness, which has high sensitivity to α-particles, and low sensitivity to γ-radiation (7) The ratio of α-sensitivity to γ-sensitivity for this chip is 34 times higher than that corresponding to a LiF chip (5 x 5 x 0.8 mm). Hence, the ultra-thin $CaSO_4$:Tm chip is most suitable for α-exposure measurements even in high γ-background environments.

The purpose of this experiment was to determine the energy-dependence of the above chips to α-particles emitted by radon daughters and thoron daughters using the same geometry used in our TLD α-particle personal dosimeter (7). To this end polycarbonate absorbers were used to change the energy of α-particles incident on the TLD chip surface.

THEORETICAL BACKGROUND

The short-lived decay products of radon (^{222}Rn), i.e., radon daughters, RaA (^{218}Po) and RaC' (^{214}Po), emit α-particles of energy, 6.00 MeV and 7.68 MeV, respectively. For a given α-potential energy concentration (Working Level), the number of 6.00 MeV α-particles and 7.68 MeV α-particles can vary, depending on the equilibrium conditions between radon and its short-lived progeny (see Table 1). The short-lived decay products of thoron, i.e., thoron daughters, ThC (^{212}Bi), and ThC' (^{212}Po), emit α-particles of energy 6.07 MeV and 8.78 MeV, respectively. For any given α-potential energy concentration, the ratio of the number of particles corresponding to these radioisotopes is constant. The theoretical values corresponding to the number of α-particles for 1 WL α-potential energy concentration are given in Table 1. From this Table it can be seen that the ratio of α-particles emitted by ^{214}Po to that of ^{218}Po varies from 1 to 16.25 and the ratio of α-particles emitted by ^{212}Po to that of ^{212}Bi is a constant value of 1.967. There is a maximum relative difference of 8.8% in the total number of α-particles of radon daughters, or thoron daughters, between the most probable equilibrium conditions in

Table 1 - Some theoretical data of radon and thoron progeny.

Progeny Products	Equilibrium Condition	α-Energy (MeV)	α-particle Count (N_1 or N_3)	$\frac{N_3}{N_1}$	Total α-particle Count ($N_{T\alpha}$)	A (%)
Radon	^{218}Po only	6.00 (^{218}Po)	9356 (N_1)	1	18712	8.8
		7.68 (^{214}Po)	9356 (N_3)			
	Equilibrium	6.00 (^{218}Po)	977 (N_1)	16.26	16850	-2.0
		7.68 (^{214}Po)	15873 (N_3)			
	X=0.43, Y=0.20	6.00 (^{218}Po)	2431 (N_1)	6.074	17197	0
		7.68 (^{214}Po)	14766 (N_3)			
Thoron	Any Condition	6.05 (^{212}Bi)	5488 (N_1)	1.967	16285	05.3
		8.78 (^{212}Po)	10797 (N_3)			

Notes:
1. 1 WL of α-potential energy concentration was referred to in calculation.
2. N_1 is the number of α-particles emitted by ^{218}Po or ^{212}Bi.
3. N_2 is the number of α-particles emitted by ^{214}Po or ^{212}Po.
4. $N_{T\alpha}$ is the total number of α-particles. $N_{T\alpha} = N_1 + N_3$.
5. A is the difference of $N_{T\alpha}$ between most probable equilibrium condition (i.e., X = 0.43 and Y = 0.2 in U.S.A. mines) and other equilibrium conditions chosen, divided by the value of $N_{T\alpha}$ of the most probable equilibrium condition.
6. [^{218}Po], [^{214}Pb] and [^{214}Po] represent the activity concentration of ^{218}Po, ^{214}Pb and ^{214}Po, respectively.
7. X = [^{214}Pb]/[^{218}Po], and Y = [^{214}Po]/[^{218}Po].

underground uranium mine atmospheres and any other equilibrium conditions. Another difference between radon daughters and thoron daughters is the energy of the α-particles. Alpha-particles emitted by ^{212}Po have an energy of 8.78 MeV which is higher than α-particles emitted by ^{214}Po. For the geometry of the dosimeter detector head (see Figure 1), the α-particles emitted by ^{218}Po, ^{214}Po, ^{212}Bi, ^{212}Po, deposited on the filter, will penetrate through the chip and only lose part of their energy in the sensitive layer of the chip. Because of a difference in the initial energy of the α-particles, these particles will lose different amounts of energy, and hence, cause different TLD response.

Determination of the α-particle energy dependence of the $CaSO_4$:Tm chip is important in order to determine the inherent error of this type of TLD chip for measuring radon daughters personal exposure in the presence of thoron daughters as is the case in some underground environments in Canada, China, the U.S.A. and other parts of the world.

EXPERIMENTAL PROCEDURE

Radon daughter sources and thoron daughter sources manufactured by Pylon Electronic Development (Ottawa), known commercially as Radon Daughter Standard RN-190 and Thoron Daughter Standard TH-190, respectively, were used to expose 0.8μm (23 mm diameter) Millipore filters (see Figure 2). The RN-190 and TH-190 sources consist of two unequal cylindrically-shaped metal halves that contain a ^{226}Ra source (RN-190) or a ^{232}Th source (TH-190). The two metal halves become an air-tight enclosure by means of a screw/clamp mechanism. Filters are placed on the base of the container (enclosure) where they are exposed to the decay products of ^{226}Ra or ^{232}Th. The filters collect these decay products simply by plate-out.

The maximum activities deposited by the RN-190 and TH-190 sources on

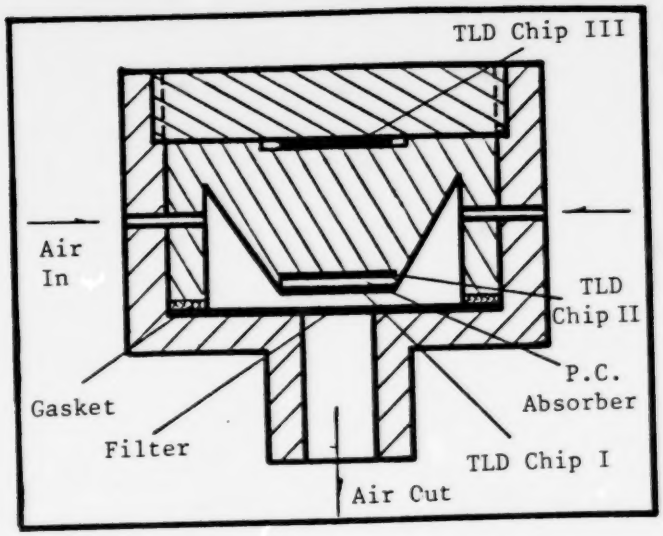


Fig. 1 - Cross-section of the dosimeter detector head.

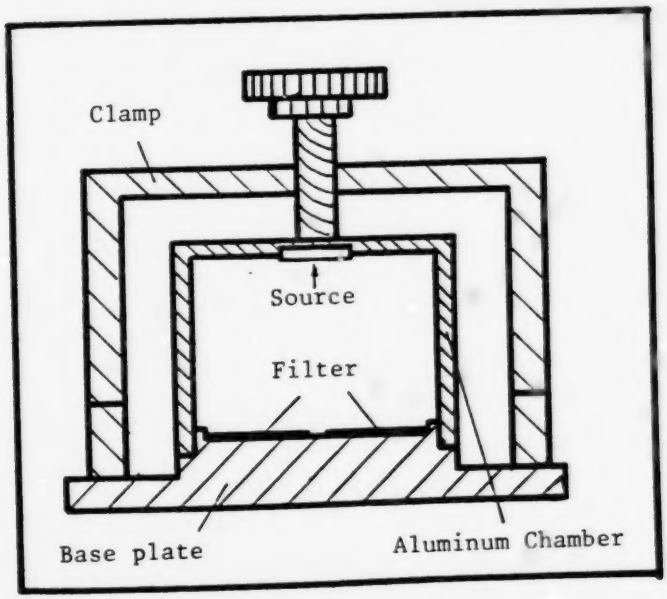


Fig. 2 - Filters placed in the standard source to collect radon or thoron progeny products.

the filters were 21000 ± 850 dpm/cm^2 and 29000 ± 2300 dpm/cm^2, respectively (dpm stands for disintegrations per minute). For the radon daughters experiment, two filters were placed side by side in the RN-190 source for a given period of time in order to deposit the same amounts of radon daughters on each filter (Figure 2). The filters were then removed simultaneously from the source; one was measured in a Multichannel Analyzer (Camberra Series 30) for a given period of time in order to determine the α-spectrum and total α-count of the filter. The other filter was placed in the detector head of the dosimeter to expose the ultra-thin $CaSO_4$:Tm chip for the same period of time. After exposure the chip was removed from the detector head and read in Model 2000A and Model 2000B TLD reading system, manufactured by Harshaw Chemical Company (U.S.A.). The TLD chip read out procedure used in this experiment was a 30-second integration period from 100^{o}C to 300^{o}C with a temperature rate of 35^{o}C/s.

The same procedure as that indicated above was followed for the thoron daughters experiment using a TH-190 source. In order to reduce the incident energy of α-particles, polycarbonate (p.c.) absorbers with different thicknesses were used to cover the radioactive filters while being measured in the Multichannel Analyzer and exposed to the TLD chips in the detector head. With this method, α-particles with incident energies different from 8.78 MeV, 7.68 MeV and 6.07 MeV were obtained (see Figure 3). The energy corresponding to α-particles emerging from polycarbonate absorbers of different thicknesses is shown in Figure 4. These energies were measured according to standard α-spectrometry calibration procedures. The response of TLD chips to the different α-particle energies indicated in Figure 4, and other energies, was investigated.

After exposure to different α-particle energies, the TLD chips were read in the Harshaw system, and their response in terms of nanoCoulomb (nC)

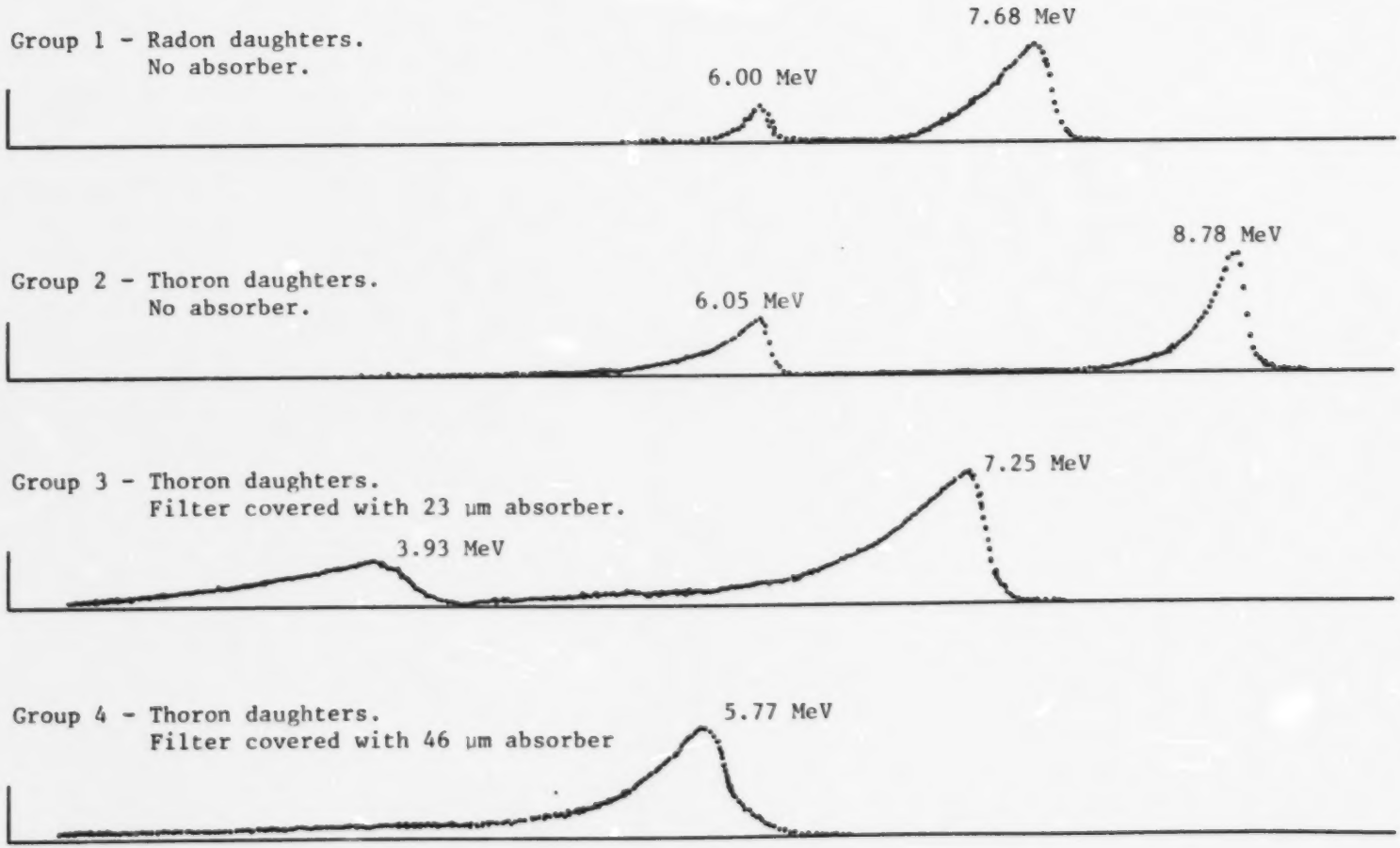


Fig. 3 - Four different energy groups of α-particles in determining energy-dependence of TL response of ultra-thin $CaSO_4 \cdot Tm$ chips.

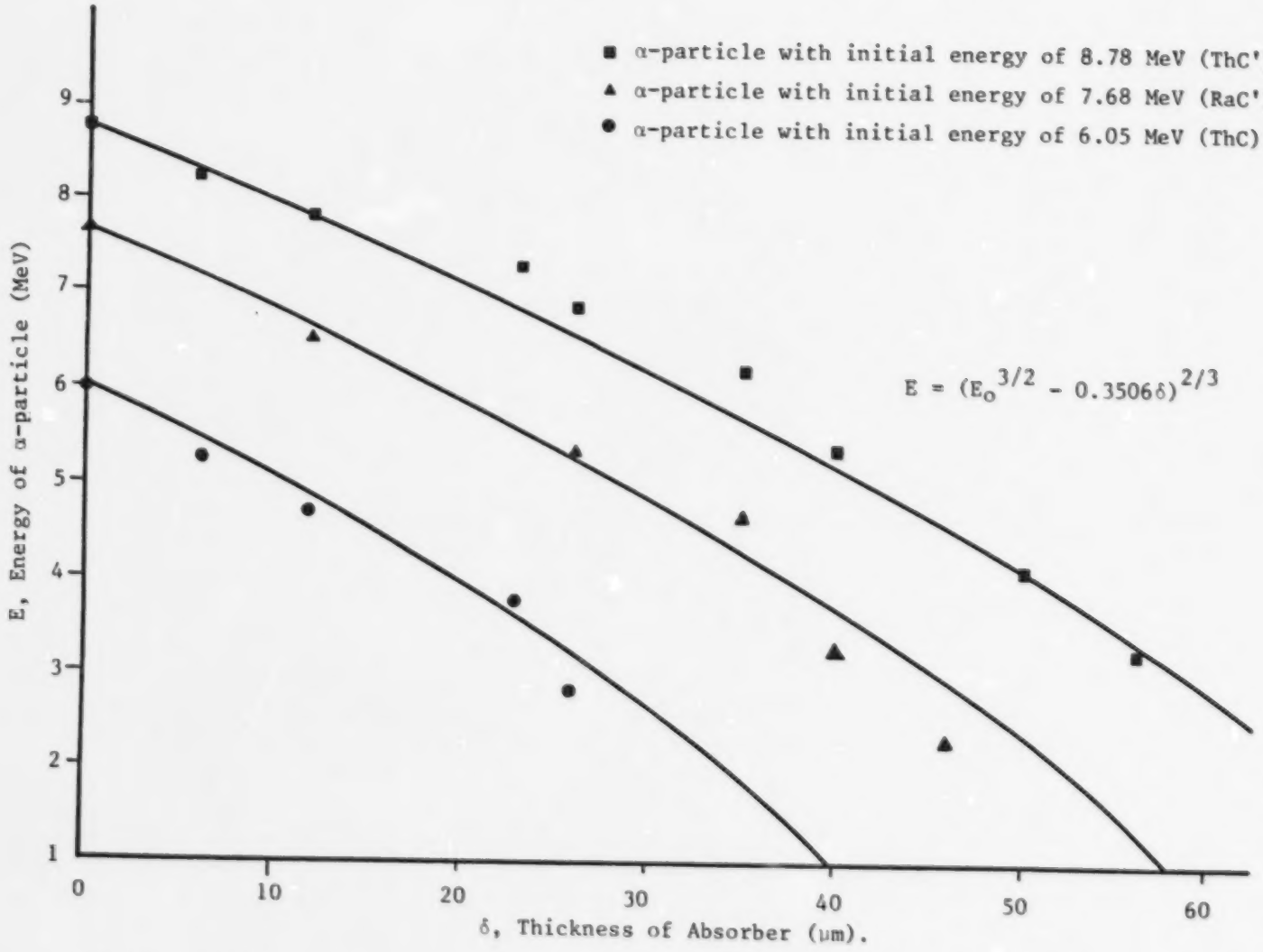


Fig. 4 - Different incident energy of α-particles emitted by ThC', RaC' and ThC deposited on the filters covered with P.C. absorber with different thickness.

per α-particle, were calculated for each energy.

RESULTS AND DISCUSSION

The results obtained have been summarized in Tables 1 and 2, and Figures 3 and 4.

Figure 3 shows the effect of polycarbonate absorbers of different thicknesses on the α-particle energy corresponding to the thoron progeny. As expected, the energy of α-particles traversing the absorber diminishes with increasing absorber thickness. Figure 3 shows that the 6 MeV α-particle from ^{212}Bi is totally absorbed by 46 μm polycarbonate.

Figure 4 shows the relationship between α-particle energy and absorber thickness. The experimental data have been fitted to a power function and the theoretical functions are shown as continuous lines. The best fitted power function to the experimental data was found to be:

$$E_\alpha = (E_o^{3/2} - 0.3506\delta)^{2/3}$$

where, δ is the thickness of the absorber in μm, and E_o is the initial energy of the α-particle, i.e., ~6 MeV, 7.69 MeV and 8.78 MeV. Figure 4 shows that less than ~40 μm is sufficient to completely absorb ^{212}Po and ^{212}Bi α-particles and less than 60 μm is needed to absorb ^{214}Po α-particles.

Tables 1 and 2 show some theoretical and experimental data, respectively. The meaning of the various columns in Table 2 is as follows.

The second column shows the α-particle energy emerging from the exposed filter alone, or from the filter and polycarbonate absorber(s), i.e., 23 μm or 46 μm, placed on the filter in order to reduce the α-particle energy.

The third column represents the ratio of the α-particle count corresponding to the high energy photopeak to the count corresponding to the low energy photopeak of the radon or thoron progeny spectrum as determined by α-spectrometry. This ratio is denoted by R, for short.

Table 2 - TLD response of ultra-thin $CaSO_4$:Tm chip to α-particles of different energy.

Energy Group	α-Energy (MeV)	R*	Average α-energy ($\bar{E}_\alpha$)	Total α-particle Count ($N_{T\alpha}$)	TLD Reading D, (nC)	$(D/N_{T\alpha})\times10^{-5}$ (nC/α-particle)	$(D/N_{\alpha T}\bar{E}_\alpha)\times10^{-6}$ (nC/MeV)
Radon Progeny (No absorber)	6.00 (^{218}Po)	6.6	7.46	367071	29.7	8.09	10.85
		8.1	7.50	460202	33.1	7.19	9.60
	7.68 (^{214}Po)	6.8	7.49	300843	20.8	6.91	9.26
		5.5	7.43	200531	15.5	7.73	10.41
		6.3	7.44	296990	23.7	7.98	10.71
						Ave: 7.58±0.51	Ave: 10.17±0.70
Thoron Progeny (No absorber)	6.05 (^{212}Bi)	1.60	7.73	1173512	82.3	7.01	9.07
		1.55	7.71	591540	44.3	7.47	9.69
		1.59	7.73	933175	68.6	7.35	9.52
	8.78 (^{212}Po)	1.59	7.73	422355	34.9	8.26	10.70
		1.60	7.73	1760244	139.3	7.91	10.24
		1.63	7.74	1105633	81.1	7.34	9.47
						Ave: 7.56=0.45	Ave: 9.78±0.59
Thoron Progeny (23 μm absorber)	3.93 (^{212}Bi)	1.20	5.75	776224	47.3	6.09	10.61
		1.38	5.86	1010861	66.7	6.60	11.27
		0.84	5.45	974835	57.4	5.89	10.81
	7.25 (^{212}Po)	1.37	5.84	807660	49.1	6.08	10.40
		1.27	5.79	939905	53.9	5.73	9.91
						Ave: 6.16±0.30	Ave: 10.60±0.50
Thoron Progeny (46 μm absorber)	5.77 (^{212}Po)	No low energy photopeak present	5.77	465610	23.8	5.11	8.86
				487422	24.8	5.09	8.82
				483171	23.4	4.84	8.39
				819699	39.3	4.79	8.31
				525718	25.0	4.76	8.24
						Ave; 4.92±0.14	Ave: 8.52±0.29

*R is the ratio of the α-particle counts corresponding to the high energy photopeak to the low energy photopeak of the radon/thoron progeny spectrum.

The fourth column represents an approximate average α-particle energy calculated over the entire spectrum. The values given are only very rough approximations as the calculations have been done using the energy corresponding to the maximum α-particle count for each photopeak and the integrated α-particle count under the corresponding photopeak.

The fifth column gives the total α-count corresponding to the entire α-spectrum, i.e., all photopeaks included. The sixth column shows the TLD reading (nC) as determined by the TLD reader.

The seventh and eighth columns represent the 'sensitivity' of the TLD chip in terms of nC/α-particle and nC/MeV, respectively. The latter values are based on the calculated average α-particle values, E_α (fourth column), and therefore, represent rough estimates subject to appreciable uncertainty.

The data in Table 2 shows that the sensitivity of ultra-thin $CaSO_4$:Tm chips is energy dependent (see 7th column), and decreases with decreasing α-particle energy. Table 2 also shows that the TLD sensitivity for unshielded, i.e., no absorber, radon progeny (~6 MeV and 7.68 MeV), and thoron progeny (~6 MeV and 8.78 MeV) is essentially the same. This can be understood because of the different decay branching ratios for the two decay series which lead to a very similar average α-particle energy in both cases. (Some additional and useful information has been summarized in Table 1.)

The fact that the sensitivity of $CaSO_4$:Tm TLD chips to radon progeny and thoron progeny, deposited on suitable substrates, e.g., 0.8 μm Millipore filters, is identical considerably simplifies the calibration of α-particle personal dosimeters using the above TLD material for field applications. This is so because the dosimeter needs only to be calibrated in radon/radon progeny atmospheres. If ultra-thin $CaSO_4$:Tm chips were strongly α-particle energy dependent, calibration of the dosimeter would be necessary under the following experimental conditions:

a) radon/radon progeny atmospheres;

b) thoron/thoron progeny atmospheres; and

c) arbitrary mixtures of a) and b).

CONCLUSIONS

The sensitivity of ultra-thin $CaSO_4$:Tm TLD chips have been examined. The data obtained can be applied to calculate a theoretical calibration factor for dosimeters using this TLD material in order to estimate personal radiation exposure under field conditions.

ACKNOWLEDGEMENT

One of us (H.L.) would like to thank the International Atomic Energy Agency (Vienna, Austria) for a fellowship to work at the Elliot Lake Laboratory (CANMET, Energy, Mines and Resources Canada) during the period April to December 1986.

REFERENCES

1. McCurdy, D.E., "Thermoluminescent dosimetry for monitoring of uranium mines"; USAEC Report COO-1500-16; Colorado State University, Fort Collins, Colorado; June 1969.

2. McCurdy, D.E., Schiager, K.J. and Flack, E.D., "Thermoluminescent dosimetry for personal monitoring of uranium miners"; Health Physics, vol 7, p 414; 1969.

3. White, O. Jr., "An evaluation of six radon dosimeters"; USAEC Health and Safety Laboratory Technical Memorandum 69-23; October 1969. "Evaluation of MIT and ORNL radon daughter dosimeters": USAEC Health and Safety Laboratory Technical Memorandum 70-3; July 1970. "Evaluation of MOD and ORNL radon daughter dosimeters"; USAEC Health and Safety Laboratory Technical Memorandum 71-17; October 1971.

4. Rock, L.R., "Measurement of employee's individual cumulative exposures to radon daughters as practised in the United States"; Personal Dosimetry and Area Monitoring Suitable for Radon and Daughter Products"; Proc. of the NEA Special Meeting, Elliot Lake, Ontario, Canada; pp 71-90; 1976.

5. Breslin, A.J., "MOD working level dosimeter"; Personal Dosimetry and Area Monitoring Suitable for Radon and Daughter Products; Proc of the NEA Special Meeting, Elliot Lake, Ontario, Canada; pp 91-100; 1976.

6. Bigu, J., Tremblay, R.J., Carson, D.W. and Townsend, M.G., "A technical evaluation of several personal α-dosimeters in Canadian uranium mines"; Division Report MRP/MRL 82-76(TR), CANMET, Energy, Mines and Resources Canada; March 1982.

7. Li, H., Pu, Q. and Zhou, J., "The development and field testing of the thermoluminescent personal α-dosimeter for measuring exposure to radon and thoron daughters"; Proc Int Conf on Radiation Safety in Mining, Toronto, Canada; 1984.